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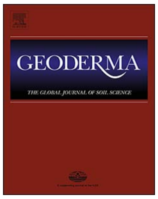

Drinking water treatment residual use in urban soils: Balancing metal immobilization and phosphorus availability

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ABSTRACT

Pollution in urban environments has raised concerns about the consumer safety of food produced by urban horticulture. Could a solid by-product of drinking water treatment provide the answer? Soil amendment with drinking water treatment residuals (DWTRs) has been shown to limit the uptake of metal contaminants, effectively immobilizing these elements within the soil; however, DWTRs possess a strong specific affinity for P and soil amendment with DWTRs can reduce P availability, thereby inducing plant P deficiencies and restricting growth. We conducted a glasshouse pot experiment to investigate the effect of alum-based DWTR amendment rate and P fertilizer placement method on *Brassica pekinensis* growth and As, Cd and P uptake under controlled conditions. Amendment of sandy loam soil A with 2–4 wt% DWTRs significantly ($p < 0.001$) reduced the Cd content of *B. pekinensis* tissues relative to untreated controls, with no significant difference in the P content of *B. pekinensis* tissues. Across all DWTR application rates examined (2–6 wt%), banded P fertilizer application resulted in 30–47% greater aboveground tissue P concentration compared to broadcast P fertilizer treatments ($p < 0.05$). Both the As and Cd content of *B. pekinensis* tissues were significantly ($p < 0.001$) reduced following 2–6 wt% amendment of sandy loam B with DWTRs. Results showed that a DWTR amendment rate of 2 wt% coupled with banded P fertilizer application resulted in the greatest *B. pekinensis* aboveground tissue biomass, along with a reduction in plant tissue Cd or As concentrations not significantly different from higher rates of DWTR amendment. Tissue P concentrations of *B. pekinensis* grown in soils amended with 2 wt% DWTRs were adequate for uninhibited plant growth. The use of DWTRs as an amendment to urban horticultural soils may provide a low-cost means of immobilizing trace metals without limiting plant uptake of applied fertilizer P. Additional study is required to quantify trace element release from DWTR-amended soils as a function of soil oxidation-reduction status.

1. Introduction

The content of trace metals such as As, Cd, Cr, Cu, Hg, Ni, Pb, Se and Zn is commonly elevated in urban soils due to human activity, with Cd, Ni and Zn in particular characterized by high soil-plant transfer coefficients (Kabata-Pendias, 2004; Hazelton and Murphy, 2011; Szolnoki and Farsang, 2013). Vehicle traffic, current and historic industry-specific activities, and the degradation of building materials are common anthropogenic sources of metals in urban areas (Wong et al., 2006). Although precise statistics concerning the proportion of the global population engaged in urban agricultural production vary, it is conservatively estimated that ≥ 100 million people make earnings directly from urban agriculture (Eigenbrod and Gruda, 2015; Orsini et al., 2013). The number of people engaged in production apart from commercial purposes is likely substantially > 100 million, highlighting the importance of urban agriculture in modern society. Vegetable cultivation dominates the urban agricultural sector and contributes most significantly to associated urban populations in a nutritional, economic and social context (Eigenbrod and Gruda, 2015; Mok et al., 2014). Given the growing world population and increase in urbanization, with more than half of the global population living in cities since 2007, urban agriculture is likely to increase, with vegetable production as the fastest growing sector due to crop nutritional value, short growth cycles and the minimal processing required after harvest. Increased urban vegetable production could increase human dietary exposure to trace metal contaminants in fresh vegetables. Soils need not be classified as metal-contaminated to yield crops containing trace metal contaminants at levels greater than human-health based

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Abbreviations: DWTR, drinking water treatment residual; SSP, single superphosphate (fertilizer); WHC, water holding capacity

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maximum permissible concentration (MPC) guidelines. For example, lettuce and broccoli grown in Australian soils containing ca. 0.2–0.3 mg Cd kg^{-1} have been found to exceed the 0.05 mg kg^{-1} Australian and New Zealand MPC for Cd in fresh vegetables (Jindasa et al., 1997). In contrast, the Australian interim urban ecological investigation level for Cd in soil is 3 mg kg^{-1}, whilst the health investigation level for residential soil is 20 mg Cd kg^{-1} (NEPM, 1999).

Drinking water treatment residuals (DWTRs) may be suitable amendments for trace metal-contaminated soils to reduce metal lability and phytoavailability. Previous work has shown that soil amendment with DWTRs reduces plant tissue concentrations of metals in ryegrass (*Lolium perenne*), tomato (*Lycopersicon esculentum*), canola (*Brassica napus*) and maize (*Zea mays*) (Elliott and Singer, 1988; Mahdy et al., 2008; Mahmoud, 2011; Silvetti et al., 2014). DWTRs are by-products of potable water treatment formed when coagulants, usually Al sulfate ($Al_2(SO_4)_3$) or Fe salts ($FeCl_3$ or $Fe_2(SO_4)_3$), are added during water purification. These salts bind suspended particles in raw water to produce a sludge, or DWTR. Precipitated DWTRs are poorly crystalline solids and are typically classified as either alum or ferric sludge, or a mixture, depending on the composition of the coagulant(s) added. DWTRs possess high cation exchange capacity and are known to effectively immobilize metals by adsorption and co-precipitation processes (Elliott and Singer, 1988; Ippolito et al., 2011; Mahdy et al., 2008). This attribute is effectively exploited in the water treatment process to remove metal contaminants and other impurities before distributing the treated water to reservoirs or through pipes for drinking purposes. The benefits from DWTRs' high metal sorption capacity can be extended to cultivated land contaminated by pollutants through use of ‘waste’ DWTRs as soil amendments. Despite their demonstrated ability to immobilize metals in soil, use of DWTRs as soil amendments is complicated by the trace element content of the DWTRs themselves, and the high affinity of DWTRs for P, an essential plant nutrient. Concerns such as the possible leaching of trace metals from DWTRs and P immobilization have hindered the widespread adoption of land application as a form of beneficial DWTR re-use to date (Lombi et al., 2010; Silvetti et al., 2014; Wang et al., 2014).

One of the primary limitations of the land application of DWTRs is their high affinity for PO_4-P (Ippolito et al., 2011). This characteristic is unfavorable for soil remediation or reclamation as soil P can become immobilized, making it unavailable to plants intended either for revegetation or cultivation purposes. Plant P deficiency has been observed in DWTR-amended soils even at low application rates due to the high surface area of the poorly crystalline Fe and/or Al oxides and (oxy) hydroxide mineral precipitates which comprise the bulk of most DWTRs, and the strong specific sorption affinity of these minerals for PO_4-P (Cox et al., 1997; Lucas et al., 1994; Rengasamy et al., 1980). Phosphorus adsorbed to poorly crystalline Fe/Al oxide and (oxy)hydroxide minerals is sparingly available to plants, and amendment of soil with DWTRs comprised primarily of Fe/Al oxide and (oxy)hydroxide minerals can induce P deficiency in subsequent crops. Although the addition of P fertilizers could correct P deficiency induced by DWTR amendment, the quantity of P fertilizer required may be cost-prohibitive (Lombi et al., 2010). Purposeful placement of P fertilizer has the potential to alleviate induced P deficiency and enable widespread productive re-use of DWTRs in horticulture by reducing contact between amended DWTRs and P fertilizer.

Examination of effective rates and techniques for application of both DWTRs and fertilizers is necessary to facilitate the beneficial re-use of DWTRs in urban horticulture. Although P phytoavailability studies with regards to DWTR application are common, few have investigated concomitant metal immobilization and P availability as a function of DWTR amendment. The present examination of both metal and P availability in soil as a function of DWTR amendment rate will facilitate elucidation of the interactions between metal and P immobilization by DWTRs and inform the optimization of DWTR amendment rates. By investigating the effects of P fertilizer application techniques in DWTR-amended soils, the present study will provide further information necessary to derive P fertilizer placement recommendations for reduced phytoavailability of trace metals and minimal P immobilization by DWTRs.

The objective of this study was to (i) evaluate the impact of the addition of DWTRs containing trace metals on Cd and As uptake by a leafy vegetable species, and (ii) to derive optimum amendment rates for DWTR application in order to maximize metal immobilization whilst maintaining sufficient P availability. The combination of DWTR sorption effects on trace metal and P phytoavailability was studied under controlled conditions using Chinese cabbage (*Brassica pekinensis* (Lour.) Rupr.) and two metal-contaminated sandy loam soils from Australia. Chinese cabbage was selected as a test species because studies have shown that metals accumulate to significantly higher levels in leafy vegetables compared to root vegetables, with Cd in particular partitioning into edible leafy portions of crops rather than fruit or storage organs (Lehoczky et al., 1998; Page et al., 1987). The effect of P fertilizer placement was also investigated to determine whether placement of P fertilizer within a dense band could minimize P sorption by DWTRs, thereby increasing plant assimilation of applied P fertilizer.

2. Materials and methods

2.1. Sample collection and preparation

Alum-based DWTRs were obtained from the North Pine Water Treatment Plant (Joyner, North Brisbane) in February 2015. Two hundred liters (200 L) of DWTR slurry, comprised of 3–6 wt% solids, was dewatered to yield dried material. Initially, the slurry settled gravimetrically in 20 L buckets with supernatant periodically removed from the surface using a submersible pump. After the volume of slurry was reduced by approximately half and additional water could not easily be removed without also removing a substantial quantity of solids, the remaining water in the mixture was evaporated in pans measuring 1 m^2 by a depth of 5 cm via exposure to ambient sunlight. Air-dried DWTR slurry was subsequently oven-dried at 105 °C for 48 h to a constant mass. Oven-dried DWTRs were either ground by hand to $<$ 1 mm for use in preliminary laboratory trials or ground to $<$ 2 mm using a mechanical grinder for use in glasshouse experiments.

Two soils were obtained for use in glasshouse experiments. Soil A, comprised of the topmost 10 cm of a Yellow Chromosol from near Bundaberg, QLD, was a sandy loam containing 0.27 mg Cd kg^{-1}. The Cd-contaminated sandy loam was retained from a 2002 study during which the soil was amended with 0.3 mg Cd kg^{-1} in the form of $CdCl_2$ (Barry and Bell, 2006). Soil B was a sandy loam soil known to be As-contaminated (13 mg As kg^{-1}), the uppermost 10–15 cm of which were collected at a former cattle tick dip site near Doubtful Creek, NSW.

2.2. Soil and DWTR analysis

Quantitative X-ray diffraction (XRD) was use to characterize the mineralogical composition of DWTRs and soils. One gram of each sample was ground to $<$ 10 μm under ethanol for 10 min in a McCrone micronizing mill and the resulting slurries oven-dried at 60 °C prior to thorough mixing with an agate mortar and pestle. Mixed samples were back-pressed into stainless steel sample holders for XRD analysis. XRD patterns were recorded with a PANalytical X'Pert Pro Multipurpose Diffractometer using Fe-filtered Co Kα radiation, 1/4° divergence slit, 1/2° anti-scatter slit, and a fast X'Celerator Si strip detector. The diffraction patterns were recorded in steps of 0.016° 2θ (theta) with a 0.4 s counting time per step and logged to data files for analysis. The commercial package TOPAS from Bruker was used to perform quantitative analysis on XRD data.

Major and trace elements within DWTRs and soils were quantified using X-ray fluorescence (XRF). Major elements were quantified by fusion XRF, wherein samples were oven dried at 105 °C before a 1 g